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## Crack tortuousity in the nacreous layer – Topological dependence and biomimetic design guideline



## Abhishek Dutta, Srinivasan Arjun Tekalur\*

Department of Mechanical Engineering, Michigan State University, East Lansing, MI 48824, USA

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#### ABSTRACT

The nacreous layer in seashells is known for two phenomenal aspects: light-weightiness and superior fracture toughness. Of a multitude of toughening mechanisms, the highly meandering nature of the crack path through its staggered architecture has been reported to contribute approximately a third of its overall toughness. In the current article, we are trying to establish the scientific rationale associated with the influence of overlap length on the crack-tip driving force from a local perspective via development of a simplified analytical model. Characteristic overlap lengths computed showed reasonable agreement with the values reported in the nacreous layer and previously published experimental data. Biomimetic design guideline obtained from the current investigation would thereby lead to development of synthetic staggered architecture materials with improved stiffness, load-transfer and toughness.

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### 1. Introduction

The nacreous layer in seashells comprises of overlapping ceramic tablets embedded in a polymeric matrix (Jackson et al., 1988; Sarikaya et al., 1990) in the form of a brick and mortar structural arrangement. In our current article, we have attempted to understand the topological principles in the nacreous layer that contributes to minimizing the crack driving force (from a local perspective) via development of a simplified analytical model. Excerpts gathered from this investigation would ultimately lead to identifying design guidelines which would promote development of synthetic staggered architecture composite materials with improved load-transfer, toughness and stiffness.

The nacreous layer is an ideal model for biomimetic inspiration (Barthelat and Espinosa, 2007; Jackson et al., 1988; Kessler et al., 1996; Kamat et al., 2000; Sarikaya et al., 1990). In addition to being both strong and stiff, it exhibits two additional predominant and promising aspects: light weight and superior fracture toughness. The toughness has been attributed to existence of a multitude of toughening mechanisms, namely tablet pull-out (Jackson et al., 1988), crack deflection through the biopolymer (Menig et al., 2000), platelet interlocks (Katti et al., 2005), presence of nanoasperities (Evans et al., 2001; Wang et al., 2001), diffusive tablet sliding (Barthelat et al., 2007) arising from tablet waviness, aragonite bridge reinforcements at interface (Song et al., 2003), re-locking of tablets (Meyers et al., 2008) due to persistent contact

of broken aragonite bridges, and synchronized deformation twinning of the nano-scale particles in ceramic bricks (Huang et al., 2011). All the aforementioned factors contribute to both interfacial strengthening and fracture resistance in varying degrees of resistance. What contributes to the superior load-transfer and energy dissipative capabilities of the nacreous layer is the meandering nature of the crack path throughout its architecture (Li, 2007). It has already been reported in the past by Jackson et al. (1988) that approximately a third of the toughness of wet nacre samples has been associated with tablet pull-out. As per Barthelat and Espinosa (2007),  $J = J_0 + (J_B + J_W)$  where  $J_0$ ,  $J_B$  and  $J_W$  correspond to intrinsic toughness, toughness associated with crack bridging and toughness contribution from the process zone. In their investigations, it was found out that  $J \approx 1.5 \text{ kJ/m}^2$ ,  $J_B = 0.021 \text{ kJ/m}^2$ ,  $J_W$  = 0.75 kJ/m<sup>2</sup>, which implies that  $J_0 \approx 0.73$  kJ/m<sup>2</sup>. Clearly it can be seen that the contribution from intrinsic toughening via crack deflection is approximately half of the overall toughness. Presence of weak interface contributes to stress redistribution around the vicinity of strain-concentration locales thereby promoting stress shielding and thus leading to toughening via crack deflection (Clegg et al., 1990; Launey and Ritchie, 2009). Note that higher the energy required to drive the crack throughout the individual microscopic unit cells, higher will be the energy dissipated and consequently superior will be the toughness of the nacreous layer on a macroscopic level. Till date, a number of literature studies have attempted to address the behavior of the staggered architecture materials on the basis of interdependence of material properties and geometrical parameters. Table 1 provides the references and the approach adopted by various researchers with reference to modeling of biological composites, in a brief manner.

<sup>\*</sup> Corresponding author. Tel.: +1 517 884 1608; fax: +1 517 884 1601. *E-mail address:* tsarjun@egr.msu.edu (S.A. Tekalur).

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#### Table 1

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Authors	Description
Jager and Fratzl (2000), Kotha et al. (2000)	Predicted modulus of staggered architecture composites along the longitudinal direction of the inclusions as a function of aspect ratio and concentration of the inclusions
de Gennes and Okumura (2000), Okumura and de Gennes (2001)	Developed an analytical model based on laminar architecture of seashells to account for the existence of weaker stress concentration ahead of the crack tip in these materials in comparison to traditional isotropic elastic materials along with an examination of the effect of differential stiffness of inclusions and interfaces
Gao et al., 2003	Developed the tension-shear-chain (TSC) model to describe the mechanics of staggered architecture composites and study the mechanical properties of these type of biological composites
Shuchun and Yueguang (2007)	Studied the interdependence of the elastic modulus of the composite and number of hierarchical levels in bone-like materials via comparison of their results against TSC model and FE (finite element) simulations
Zhang et al. (2010)	Investigated the effect of platelet distribution (regular, stairwise, random) on stiffness, strength, failure strain and energy absorption capability in materials exhibiting staggered architecture
Zhang et al. (2011)	Used a quasi-self-similar hierarchical model to comprehend the existence of an optimal number of hierarchical levels in biological composites
Liu et al. (2011)	Provided analytical expressions for displacement and stress fields in staggered nanocomposite structures under static loading conditions under uniaxial tension
Barthelat and Rabiei (2011)	Developed a micromechanical model taking into consideration the effect of toughening associated with the process zone

The nacreous layer can be visualized as translation of unit cell structure in 2-dimensions comprising of overlapping mineral tablets having distinct overlap and core regions. The overlap region is responsible for inter-mineral load transfer via shear deformation of the polymer. One might consider what could possibly be the reason associated with the existence of specific overlap length in the nacreous layer in seashells. Analytical models and numerical simulations have been developed and certain criterion been proposed to address this aspect as listed in Table 2 below.

An extensive set of literature exists on modeling and experimentation of adhesively bonded structures under quasi-static rates of loading. For instance, Oplinger (1994) furthered the Goland and Reissner analysis by decoupling adhesive layer deflections in two halves of the joint in bending deflection analysis. Investigation by Tsai et al. (1998) developed theoretical solutions which predicted adhesive shear stress distribution in single- and double-lap joints by taking into consideration the assumption of linear shear stress distribution through adherend thickness. Predictions by theoretical model were in good agreement especially for the case of fiber composite adherends. Investigations by Chiang and Chai (1998) focused on modeling and obtaining full-field, plane-strain elastoplastic solutions for adhesively bonded structures with an interfacial crack deforming in shear, where the adhesive was modeled as elastic perfectly plastic material and adherend considered as both rigid and compliant. It was observed that during the initial stages of loading, the plastic deformation of crack-tip was shear dominated along the interface irrespective of existing triaxiality. Chai and Chiang (1998) provided quantitative insight into the

mechanics of other failure modes observed during experimentation. Microdebond growth occurring ahead of the crack tip was stipulated to be controlled by bond-normal tensile stress whereas tensile stresses (hydrostatic in nature) were considered responsible for kinking ahead of the crack tip which subsequently promotes crack arrest. All these and other failure modes are activated under large strains, which manifests the importance of plasticity in the fracture of polymeric joints. Kafkalidis and Thouless (2002) used cohesive-zone approach in lap-joint analysis to take into account cohesive properties of the interface and plastic deformation of the adherend. Luo and Tong (2004) presented analytical models for analyzing thin and thick adhesive bonded structures via taking into consideration linear variations of displacement components across the adhesive thickness, higher order displacement theory (HODT), longitudinal strain and the Poisson's effect of the adhesive. The predicted shear and peeling stress distribution was compared against classical Goland and Reissner theory followed by numerical validation. Tsai and Morton (2010) used full-field Moire interferometry to study in-plane deformations of the edge surface of joint overlaps and compared it with shear-lag solution and linear elastic 2D finite element model for double lap joints with unidirectional and quasi-isotropic composite adherends.

Since the current scenario under consideration is representative of an interface fracture problem, on a conservative approach it is opportune to consider that an interface is as much fracture resistant as the least tough participating component in the system. As per Griffith criterion, cracks will initiate and delamination or cohesive failure will occur in the unit-cell once the crack driving force (rate

#### Table 2

Recent advances on investigating existence of characteristic overlap length in staggered architecture biological composites by various researchers.

Authors	Description
Gao (2006)	Developed the principle of flaw tolerance to address the aspect of choice of nanometer sized inorganic reinforcements in natural structural composites. Additionally, genetic algorithm (GA) optimization scheme was employed to illustrate the reason behind evolution of staggered microstructural arrangement in an attempt to optimize stiffness and toughness for mechanical support and flaw tolerance
Chen et al. (2009)	Under static rates of loading, the existence of characteristic length was attributed to attainment of efficient stress transfer in staggered biocomposites
Wei et al. (2012)	Under static rates of loading, the existence of unique overlap length in biological composites was attributed to an attempt to optimize both strength and toughness frontiers in staggered architecture biological composites. Note that, the toughness in their investigations was defined in terms of elastic strain energy density
Dutta et al. (2013)	From the perspective of impact rates of loading, existence of optimal overlap length was attributed to an attempt to obtain maximum shear transfer efficiency
Barthelat et al. (2013)	Developed a criterion to predict reinforcement failure mode (either via pull-out or tablet fracture) in staggered architecture composite taking into consideration presence of a flaw in the reinforcement
Dimas and Buehler (2013)	Used molecular mechanics based model to show that by utilizing elastic constitutive laws for the matrix and tuning the matrix–reinforcement interactions, it is possible to achieve superior toughness in staggered architecture composite systems. They proposed that stiffness ratio of the constituents in the linear elastic regime is responsible for controlling deformation and fracture mechanism in biological composite systems

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